The document lists the comments (written here in small font slanted) and our responses to **Reviewer #2** .

We kindly appreciate the extensive review of our manuscript. The comments were great guidelines to improve the paper. Note that in the *Specific comments* some comments are only answered with DONE if the corrections were directly applied to the manuscript but all the aspects were addressed if not otherwise stated in an answer.

General comments:

The manuscript of Hochstaffl et al. present an intercomparison of different CH4 retrieval approaches for airborne hyperspectral measurements during an overflight of the HySpex instrument over a known CH4 emission source in Poland. Gives a short overview over the different techniques and present the retrieved maps for specific scene which are then discussed. CH4 retrievals from hyperspectral measurements is a quickly evolving field and there is merit in intercomparing different retrieval methods based on real, measured spectra.

The manuscript is in principle relevant and suitable for Atmos. Meas. Tech. but there are several shortcoming that limit its value. The manuscript is somewhat dominated by the method section while the discussion could be more detailed. In this section, a lot of material is squeezed in which makes it hard to read and I am not sure how much the reader gets out of it, especially since variables are not always well defined or used in consistent manner. The presented analysis is purely qualitative, very brief and limited to a single scene. The lack of ground truthing means that it is not possible to tell which method is best and thus the study is limited to comparisons against each other which then should be done more rigorously.

The revised manuscript has successfully addressed the issues that were identified in the initial feedback. The authors have made an effort to balance the method section with a more concise and well-structured discussion and conclusion. The material has been rearranged in a way that hopefully makes it easier to read and understand, and the variables are now well-defined and consistently used throughout the manuscript. The analysis has been expanded to include multiple scenes (scene 09 and scene 11). The authors has taken into account the feedback fo do a comparison of the method against each other more rigorously by adding two additional sections to the manuscript: Sec. 3.3 titled *Statistical significance of results* and Sec. 3.4 *Errors and correlations*. Both analyze the retrieval results in a more rigorous manner.

Main comments are:
Several figures need to be improved and labelled consistently (see detailed comments below). Especially the map of retrieved methane should be shown for the same number of cross and along track pixels so that results can be compared. Also, give results in CH4 for all methods and not as scalding factors.

The figures were improved so that they are labelled consistently, making it easier for readers to understand and compare the results. In particular, the maps of retrieved methane are now shown for the same number of cross- and along-track pixels, allowing for a easier comparisons. Moreover, results are now consistently presented in terms of CH4 mole fractions for all methods, rather than as scaling factors etc. The improvements should definitely enhance the readability of the study.

The analysis should be more quantitative. In the discussion, you state that the MF method can be used as reference, so lets also do this. You can apply simple methods to identify a plume (eg thresholding) which will then allow you do contrast inferred plume shapes. Also, I would like to see correlation plots of CH4 enhancements between different methods for example for pixels within the plume (eg defined according to the the MF method).

The revised manuscript has successfully addressed the issue of the lack of quantitative analysis. The authors has applied the t-test method (Sec. 3.3) on the rerieval results from different methods to identify the plume and compute statistics upon. Additionally, the author has provided correlation plots of CH4 results between different methods. These changes should allow readers to better understand the relationships between the different methods.

In course of the revision of the manuscript we decided to not use the MF method as a benchmark but do initially not prefer a method and simply compare each against all others. It was found

that the nonlinear covariance weighted BIRRA retrieval yields best results in terms of contrast and with the smallest statistical uncertainties.

Also, check all equations and ensure that all variables are fully defined and used consistently. For example, beta is used to describe 3 different variables.

In the revised manuscript the issue of inconsistent variable is resolved. All variables are fully defined and used consistently throughout the manuscript. Hence it should be easier now to follow the methodology and the subsequent analysis. If the authors overlooked a case we kindly ask to report it.

Specific comments:

Page 2, line25: important greenhouse gas -¿ important anthropogenic greenhouse gas

DONE

Page 2 line 32: fossile -¿ fossil

DONE

Page 2 line 38: besides satellites, there are the global in-situ surface networks

The networks Global Atmosphere Watch (GAW) and Integrated Carbon Observation System (ICOS) were added

Page 2 , line 55: smaller emitting area. This does not refer to IR so it is reflecting rather than emitting.

Corrected this to . . . loss of photons caused by the smaller ground pixels . . .

Page 3, line 61: add reference for CHIME, eg. Rast et al., IEEE, 2021

DONE

Page 3, lines 60-63: add MethaneSat for completeness

DONE

Page 3, line 75: slowly varying part is also from scattering

DONE

Page 3, line 81: This study compares various retrieval schemes . . . -¿ please add the goal of study

We added a statement before the methodologs section.

Page 4, section 2.1: I suggest to add a table with the key instrument parameters for the HySpex instrument

DONE

Page 4, line 96: in detail in (IMF) -¿ missing reference ?

Corrected

Page 4, figure 1b: The fonts on the figure are too small and can not be read in a hardcopy.

Adapted the figure.

Page 4, line 97: in the following chapters -¿ in the following sections

Not applicable anymore.

Page 4, line 101: . . . and if so how accurate . . . -¿ I don't think that this is addressed in this manuscript ?

Reformulated since an accuracy assessment would required some sort of truth reference.

Page 4, line 108: seen in Image 1a, -¿ Figure 1a

DONE

Page 5, line 109: wind data for the USCB area -¿ This needs to be defined separately to the definition in the abstract. However, I don t see the need to introduce this acronym as it is not used anywhere else.

It is used three times and we decided to keep the acronym but introduce it only in the text not in the abstract.

Page 5, line 119: 967-2496 nm (4005-10338cm-1). Throughout the manuscript at some places wavelength and and at other wavenumber is used. Since wavenumber is primarily used, I suggest to use consistently wavenumber throughout the manuscript and give everything in wavenumber and not wavelength.

In accordance with comments by other reviewers, it was decided to introduce wavelength but keep wavenumbers in particular for the naming of the retrieval windows since e. g. absorption lines in molecular spectroscopy databases are and hence cross sections are usually given on wavenumber grids.

Page 6, figure 2: a) please make fonts larger and lines thicker. In a hardcopy this figure is hard to read. Also, can you give a reference for the albedo data

This figure was compiled from another person and will be updated in the final version before possible publication.

b) Use either 'wavenumber' or 'wavenumbers' for the x-axis. Use either round or square brackets to give units. Label the y-axis with the shown parameter (not only units).

Remove the 2 extra digits on the x-axis labels. Caption: CH4 -¿ CH4

I don't think that adding the vertical lines to indicate spectral pixels adds value and neither does overplotting all spectra into the figure. I suggest to remove the lines and show a mean spectrum and a standard deviation. You could add the one spectrum with the outlier.

The figure was adapted according to the reviewers suggestions.

Page 6, line 131, see absorption from methane's 2v3 band around 6000cm-1 -¿ where do I see this. Can you label this in the figure ?

This statement was removed for the sake of clarity and since it is not clear whether the feature represents the methane's 2v3 band absorption.

Page 7, line 136: under clear sky conditions (cloud free) -¿ also scattering free in general

DONE

Page 7, eq. 1: ds needs to be removed in the sum of the first equation. define all variables including tau, nu, p, T, s and m

DONE

Page 7, lines 145-150: I don't see how the extract on aerosol optical properties is relevant. I suggest to remove this and simply refer to a textbook. The use of wavenumber of wavelength in this section is unnecessary. K_aer: give units.

The paragraph was removed and a reference with the formulas therein added.

Page 7, line 153: composed by pure -¿ composed of pure

Not applicable anymore.

Page7 , lines 154-155: define z, also I don't see the need to use zmol, zsc instead of simply z.

DONE

Page 7, eq.5, define tau_bg and tau_pl. What is alpha here ?

Variable tau_bg is now explained and tau_pl changed to tau.

Page 8, line 161: The CH4 background as well as the CO2 initial guesses -¿ The CH4 background profile as well as the CO2 background profile

DONE

Page 8, figure 3: BoA, TOA -¿ BOA, TOA; Al least for CO2 and CH4, a mixing ratio profile would be more meaningful

Not applicable anymore.

Page 9,line 186: is SRF different to ISRF ? This are examples for the many acronyms introduce but not used in the manuscript.

The use now ISRF throughout the manuscript.

Page 10, figure 4: remove unnecessary digits on x label. Thicker lines in panel b would be helpful

DONE

Page 10, line 194: Jacobian matrix -¿ define Jacobian

DONE

Page 11, Figure 5: give the definition of alpha and r in caption . Remove unnecessary digits

This plot was removed for brevity and as other authors deemed it not necessary.

Page 11, line 202: the converged spectrum-¿ the converged spectrum I

DONE

Page 11, line 206: from the diagonal elements -¿ from the square root of the diagonal elements

DONE

Page 12: beta is already used as Angstrom coefficient on page 7. Please use another variable name here.

DONE

Page 12, eq. 11: define meaning of y hat.

The variable was substituted by $\vec{y}$ for consistency with the description of other methods.

Page 13, define alpha tilde is.

The variable is explained. However, note that in course of the revision it was decided to formulate the light path correction according to literature mentioned in Sec. 2.3.2 and disregard the difference in methane and carbon dioxide in the lowest layer.

Page 13, line 231: What is a scene average scaling factor. I don't believe that the given references apply such a scene average scaling factor.

The aim was to provide references that also account for scattering by the proxy method. In our setup the co-retrieval of CO2 with methane was problematic hence this approach was chosen.

Page 13, eq. 12: usually the CH4 to Co2 ratio is multplied by a 'known' CO2 profile. If you use the co2 scaling factor directly as a correction for light path modificatiins then you assume that the CO2 profile is perfect and that alpha is 1 in absence of scattering.

This is true and the assumption is now explicitly stated in the manuscript.

Page 14, line 256: the (saturated, see . . . -¿ the saturated (see . . .

DONE

Page 14: eq 14: what is the meaning of the up and down arrows in the optical depth. Why is there a beta factor in the

Taylor expansion why is not in the exp function. Also, how is beta defined. Note that beta is used already twice with other meanings.

Arrows are representing the up- and downward paths but the notation was changed to simply τ. Also β is not used anymore in this context.

Page 14, lines 261-261: M and N is now used in capital but was used before in small letters m and n (page 11)

Capital M now only represents measurements and m is a variable for a certain species (molecule).

Page 14, line 269, condition number of 885 -¿ Please put this in some context. Which condition number is sufficient and which not.

The discussion on condition numbers was removed as other reviewers question if this part is necessary. Moreover the nonlinear fits are now made with the simple state vector $\vec{x} = (\alpha, r_0, r_1, r_2)$.

Page 15, eq. 16: isn't (J - mu) the target spectrum t . If so, then use t in equation.

DONE

P15, l284: here tau and beta is defined which would already be needed with eq. 14

This has been resolved but some quantities such as e. g., α are specific to the used methodology, however, it should always represents the enhancement factor.

P 15, eq. 18: is t here now the Jacobian. Jacibas so far is called J while t has been used as target spectrum in section 2.5.1.

The variable J is now only used for the Jacobian matrix in the nonlinear fit and t is used for the target signature in the MF and SVD schemes.

Page 16, figure 7: What does stand. rad. mean? State in caption that u1-u4 are singular vectors ?

DONE

Page 17, eq. 21: meaning of beta ? I assume this is again different to the 3 previously define beta's?

The variable β now only represents the aerosol exponent.

Page 17, line 343: retrieal's -¿ retrieval's

DONE

Page 18, line 351: The state vector x = (3m,3r) was found to be robust toward low SNR . . . -¿ what do you mean by robust?

Findings from the analysis with simple simulated measurements were found to be not applicable to the actual HySpex observations for the majority of cases so that this analysis was removed from the manuscript.

Page 18: lines 348: changing the resolution will typically also change the SNR

For the sake of brevity and because only a simple state vector $\vec{x} = (\alpha, r_0, r_1, r_2)$ was finally used in the actual retrievals, the section on the feasibility of state vecotrs was removed.

Page 18, table 1: condition numbers need to be put into context.

This analysis of condition numbers for different state vectors was deleted for the above mentioned reason.

Page 18, lines 354: use wavenumber here and not wavelength so that it is consistent

DONE

Page 20: figure 8: can you add figure with retrieved surface reflectivity.

Since the surface reflectivity coefficients are regarded an effective parameter which captures all smooth components it was decided to not devote them separate plots. In the error plots the spectral residual norm is a proxy for the albedo which is usually larger over bright areas.

Page 21, Figure 9 : make figure the same along track and across track range as figure 8. Please make all the plume figures the same range for comparability. Also, give CH4 on maps and not the scaling factors (figure 13 and 14, 15).

DONE

Page 22, figure 10: Hard to see anything. I suggest to plot only lines instead of lines+symbols.

Not applicable anymore. Plot was removed as the 2D-maps contain this information and provide a more complete picture.

Page 26, lines 424: The relative enhancement is slightly better represented in Fig. 14a.-¿ I don't think you can tell what is better as you don't know the truth.

A more rigorous statistical assessment for plume identification and correlations between different retrieval methods is now performed.

Page 27: The method yields consistent results for both spectral intervals. -¿ results shown in Figure 16 are very different. In which way are they consistent ?

Consistent with respect to the background since retrieval methods are expected to yield values close to the initial guess for background pixels (not impacted by the plume), rather independent of the spectral interval used.

Page 28, line 444, should Figure 16 have 3 rows for zero, 1 and second order ?

The analysis was reduced to zero and second order polynomials in the 6K window.

Page 29, Figure 16: can you use same colour scale and format to increase the comparability ?

DONE

Page 30: A validation from independent measurements is hence outside the scope of this study -¿ A validation from independent measurements is outside the scope of this study

DONE

Page 30, lines 461: the results from the well established MF method can be considered some sort of verification -¿ what is the justification for this. Is there a reference that can be used in support ? Also, the results from MF are not used as reference for the analysis.

In course of the revision of the manuscript we decided to not use the MF method as a benchmark but do initially not prefer a method.

Page 30, line 463: and the results agreed well with $\approx 3\,\%$*. -¿ where is this shown ?*

The spectral unmixing is now only briefly described as a purely data-driven approach in the SVD section. However, it is not shown and put into perspective in the actual analysis of the manuscript. Moreover, it would require another method to be introduced and assessed.

Page 30, line 467: was found to be the most sensitive method for the detection of enhanced methane -¿ how is this conclusion drawn ?

This statement is now underpinned by the statistical analysis in Sec.3.3.

Page 30,line 478: and potentially quantified from HySpex -¿ you have not shown this.

With respect to errors which is now consolidated in Sec. 3.3 and 3.4.

Page 31: line 494: agree well on the plume's shape. -¿ this is not shown in the manuscript

A more quantitative analysis of the plume's shape is now inferred from a t-test (see Sec. 3.3).

Page 30, conclusion: can you discuss if the findings from this study are only applicable to Hyspex or also to wider range of hyperspectral sensors for example on satellites.

DONE

With best regards,

Philipp Hochstaffl and co-authors